

CLAIMS

We claim:

1. A method of preparing a liquid, bromine-containing solution, comprising:
 - a. combining a complexing agent, hydrogen peroxide, and anhydrous
5 hydrogen bromide gas; and
 - b. adding an alkaline source.
2. The method of Claim 1, wherein said complexing agent is sulfamic acid, and
further, wherein the mole ratio of said sulfamic acid to said anhydrous
hydrogen bromide is between about 0.75:1 and about 1.5:1.
- 10 3. The method of Claim 2, wherein said alkaline source is selected from the group
consisting of alkali metal carbonate, earth alkali metal carbonate, alkali
metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth
alkali metal oxide, alkali metal hydroxide, and earth alkali metal
hydroxide.
- 15 4. The method of Claim 3, wherein said alkaline source is an alkali metal hydroxide,
and further, wherein said alkali metal hydroxide is 50% sodium hydroxide
solution.
5. A bromine-containing liquid prepared by the method of Claim 1.
6. A method of preparing a liquid, bromine-containing solution, comprising:
 - a. combining a source of bromide ions, hydrogen peroxide, and a
20 complexing agent; and
 - b. adding an alkaline source.

7. The method of Claim 6, wherein said source of bromide ions is aqueous hydrogen bromide and said complexing agent is sulfamic acid, and further, wherein the mole ratio of said sulfamic acid to said aqueous hydrogen bromide is between about 0.75:1 and about 1.5:1.
- 5 8. The method of Claim 6, further comprising after step a but before step b, adding a solid halogenating agent and another alkaline source, and then conducting a solid-liquid separation.
9. The method of Claim 8, wherein said solid halogenating agent is an organic halogenating agent.
- 10 10. The method of Claim 9, wherein said organic halogenating agent is selected from the group consisting of trichloroisocyanuric acid, sodium dichloroisocyanurate, sodium dichloroisocyanurate dihydrate, potassium dichloroisocyanurate, dichloroisocyanuric acid, trichloromelamine, N-chloro-*p*-toluenesulfonamide, N-chloromethanesulfonamide, N-chlorosuccinimide, N,N'-1,3-bromochloro-5,5-dimethylhydantoin, N,N'-1,3-bromochloro-5-ethyl-5-methylhydantoin, 1,3-dibromo-5,5-dimethylhydantoin, 1,3-dichloro-5,5-dimethylhydantoin, and N-bromosuccinimide.
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11. The method of Claim 10, wherein said organic halogenating agent is trichloroisocyanuric acid.
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12. The method of Claim 8, wherein said solid halogenating agent is an inorganic halogenating agent.

13. The method of Claim 12, wherein said inorganic halogenating agent is selected from the group consisting of calcium hypochlorite, lithium hypochlorite, and magnesium hypochlorite.
14. The method of Claim 13, wherein said inorganic halogenating agent is calcium hypochlorite.
15. The method of Claim 8, wherein said alkaline sources are selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.
16. The method of Claim 15, wherein said alkaline sources are alkali metal hydroxides, and further, wherein said alkali metal hydroxides are 50% sodium hydroxide solution.
17. A bromine-containing liquid prepared by the method of Claim 6.
18. A method of preparing a liquid bromine- and chlorine-containing solution, comprising:
- combining a source of bromine ions, hydrogen peroxide and a complexing agent;
 - adding a first alkaline source and a solid chlorinating agent;
 - conducting a solid-liquid separation; and
 - adding a second alkaline source.

19. The method of Claim 18, wherein the molar equivalent of the combination of said hydrogen peroxide and said solid chlorinating agent is in excess of the molar equivalent of said bromide ions.
20. The method of Claim 18, wherein said source of bromide ions is aqueous hydrogen bromide and said complexing agent is sulfamic acid, and further, wherein the mole ratio of said sulfamic acid to total halogen is between about 0.75:1 and about 1.5:1.
21. The method of Claim 18, wherein said solid chlorinating agent is an organic chlorinating agent.
22. The method of Claim 21, wherein said organic chlorinating agent is selected from the group consisting of trichloroisocyanuric acid, sodium dichloroisocyanurate, sodium dichloroisocyanurate dihydrate, potassium dichloroisocyanurate, dichloroisocyanuric acid, trichloromelamine, N-chloro-*p*-toluenesulfonamide, N-chloromethanesulfonamide, N-chlorosuccinimide, N,N'-1,3-bromochloro-5,5-dimethylhydantoin, N,N'-1,3-bromochloro-5-ethyl-5-methylhydantoin, and 1,3-dichloro-5,5-dimethylhydantoin.
23. The method of Claim 22, wherein said organic chlorinating agent is trichloroisocyanuric acid.
24. The method of Claim 18, wherein said solid chlorinating agent is an inorganic chlorinating agent.

25. The method of Claim 24, wherein said inorganic chlorinating agent is selected from the group consisting of calcium hypochlorite, lithium hypochlorite, and magnesium hypochlorite.
26. The method of Claim 25, wherein said inorganic chlorinating agent is calcium hypochlorite.
27. The method of Claim 18, wherein said first and said second alkaline sources are selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.
28. The method of Claim 27, wherein said first and said second alkaline sources are alkali metal hydroxides, and further, wherein said alkali metal hydroxides are 50% sodium hydroxide solution.
29. A bromine- and chlorine-containing liquid prepared by the method of Claim 18.
30. A method of preparing a liquid, bromine-containing solution, comprising:
- combining a source of bromide ions, a complexing agent, and a solid halogenating agent;
 - conducting a solid-liquid separation; and
 - adding an alkaline source.
31. The method of Claim 30, wherein said source of bromide ions is sodium bromide solution and said complexing agent is sulfamic acid, and further, wherein the mole ratio of said sulfamic acid to said sodium bromide solution is between about 0.75:1 and about 1.5:1.

32. The method of Claim 30, wherein said solid halogenating agent is an organic halogenating agent, and further comprising before step b, adding another alkaline source.
33. The method of Claim 32, wherein said organic halogenating agent is selected from the group consisting of trichloroisocyanuric acid, sodium dichloroisocyanurate, sodium dichloroisocyanurate dihydrate, potassium dichloroisocyanurate, dichloroisocyanuric acid, trichloromelamine, N-chloro-*p*-toluenesulfonamide, N-chloromethanesulfonamide, N-chlorosuccinimide, N,N'-1,3-bromochloro-5,5-dimethylhydantoin, N,N'-1,3-bromochloro-5-ethyl-5-methylhydantoin, 1,3-dibromo-5,5-dimethylhydantoin, 1,3-dichloro-5,5-dimethylhydantoin, and N-bromosuccinimide.
34. The method of Claim 33, wherein said organic halogenating agent is trichloroisocyanuric acid.
35. The method of Claim 32, wherein said alkaline sources are selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.
36. The method of Claim 35, wherein said alkaline sources are alkali metal hydroxides, and further, wherein said alkali metal hydroxides are 50% sodium hydroxide solution.

37. The method of Claim 30, wherein said solid halogenating agent is an inorganic halogenating agent.
38. The method of Claim 37, wherein said inorganic halogenating agent is selected from the group consisting of calcium hypochlorite, lithium hypochlorite, and magnesium hypochlorite.
39. The method of Claim 38, wherein said inorganic halogenating agent is calcium hypochlorite.
40. The method of Claim 37, wherein said alkaline source is selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.
41. The method of Claim 40, wherein said alkaline source is an alkali metal hydroxide, and further, wherein said alkali metal hydroxide is 50% sodium hydroxide solution.
42. A bromine-containing liquid prepared by the method of Claim 30.
43. A method of preparing a liquid bromine- and chlorine-containing solution, comprising:
- combining a source of bromide ions, a complexing agent, and a solid chlorinating agent;
 - conducting a solid-liquid separation; and
 - adding an alkaline source.

44. The method of Claim 43, wherein said source of bromine ions is sodium bromide solution and said complexing agent is sulfamic acid, and further, wherein the mole ratio of said sulfamic acid to said sodium bromide solution is between about 0.75:1 and about 1:5.1.
- 5 45. The method of Claim 43, wherein a molar excess of said solid chlorinating agent to said bromide ions is employed.
46. The method of Claim 45, wherein said solid chlorinating agent is an organic chlorinating agent, and further comprising before step b, adding another alkaline source.
- 10 47. The method of Claim 46, wherein said organic chlorinating agent is selected from the group consisting of trichloroisocyanuric acid, sodium dichloroisocyanurate, sodium dichloroisocyanurate dihydrate, potassium dichloroisocyanurate, dichloroisocyanuric acid, trichloromelamine, N-chloro-*p*-toluenesulfonamide, N-chloromethanesulfonamide, N-chlorosuccinimide, N,N'-1,3-bromochloro-5,5-dimethylhydantoin, N,N'-1,3-bromochloro-5-ethyl-5-methylhydantoin, and 1,3-dichloro-5,5-dimethylhydantoin.
- 15 48. The method of Claim 47, wherein said organic chlorinating agent is trichloroisocyanuric acid.
- 20 49. The method of Claim 46, wherein said alkaline sources are selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide,

earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.

50. The method of Claim 49, wherein said alkaline sources are alkali metal hydroxides, and further, wherein said alkali metal hydroxides are 50% sodium hydroxide solution.
51. The method of Claim 45, wherein said solid chlorinating agent is an inorganic chlorinating agent.
52. The method of Claim 51, wherein said inorganic chlorinating agent is selected from the group consisting of calcium hypochlorite, lithium hypochlorite, and magnesium hypochlorite.
53. The method of Claim 52, wherein said inorganic chlorinating agent is calcium hypochlorite.
54. The method of Claim 51, wherein said alkaline source is selected from the group consisting of alkali metal carbonate, earth alkali metal carbonate, alkali metal bicarbonate, earth alkali metal bicarbonate, alkali metal oxide, earth alkali metal oxide, alkali metal hydroxide, and earth alkali metal hydroxide.
55. The method of Claim 54, wherein said alkaline source is an alkali metal hydroxide, and further, wherein said alkali metal hydroxide is 50% sodium hydroxide solution.
56. A bromine- and chlorine-containing liquid prepared by the method of Claim 43.
57. A method of preparing a bromine-containing solid, comprising:

- a. combining a bromine compound in the oxidation state of -1, hydrogen peroxide, and a complexing agent;
 - b. adding an alkaline source; and
 - c. promoting crystallization of a bromine-containing solid.
- 5 58. The method of Claim 57, further comprising after step a but before step b, adding a solid halogenating agent and another alkaline source, and then conducting a solid-liquid separation.
59. The method of Claim 58, wherein said solid halogenating agent is an organic halogenating agent.
- 10 60. The method of Claim 58, wherein said solid halogenating agent is an inorganic halogenating agent.
61. The method of Claim 57, further comprising after step c, recovering said bromine-containing solid.
62. The method of Claim 61, wherein said bromine-containing solid is selected from
- 15 the group consisting of the alkali metal salt of hydrated N-bromosulfamate and the earth alkali metal salt of hydrated N-bromosulfamate.
63. The method of Claim 61, further comprising after recovering said bromine-containing solid, dehydrating said solid.
64. The method of Claim 63, wherein said dehydrated bromine-containing solid is
- 20 selected from the group consisting of the alkali metal salt of anhydrous N-bromosulfamate and the earth alkali metal salt of anhydrous N-bromosulfamate.

65. A method of preparing a bromine-containing solid in equilibrium with its saturated solution, comprising:
- a. combining a bromine compound in the oxidation state of -1, hydrogen peroxide, and a complexing agent;
 - 5 b. adding an alkaline source;
 - c. promoting crystallization of a bromine-containing solid; and
 - d. recovering a slurry of said bromine-containing solid in equilibrium with its saturated solution.
66. The method of Claim 65, further comprising after step a but before step b, adding
10 a solid halogenating agent and another alkaline source, and then conducting a solid-liquid separation.
67. The method of Claim 66, wherein said solid halogenating agent is an organic halogenating agent.
68. The method of Claim 66, wherein said solid halogenating agent is an inorganic
15 halogenating agent.
69. The method of Claim 65, wherein said bromine-containing solid is selected from the group consisting of the alkali metal salt of hydrated N-bromosulfamate and the earth alkali metal salt of hydrated N-bromosulfamate.
70. A method of preparing a bromine-containing solid, comprising:
- 20 a. combining a source of bromide ions, a complexing agent, a first alkaline source, and a solid, organic halogenating agent;
 - b. conducting a solid-liquid separation;
 - c. adding a second alkaline source; and

d. promoting crystallization of a bromine-containing solid.

71. The method of Claim 70, further comprising after step d, recovering said bromine-containing solid.

72. The method of Claim 71, wherein said bromine-containing solid is selected from the group consisting of the alkali metal salt of hydrated N-bromosulfamate and the earth alkali metal salt of hydrated N-bromosulfamate.

73. The method of Claim 71, further comprising after recovering said bromine-containing solid, dehydrating said bromine-containing solid.

74. The method of Claim 73, wherein said dehydrated bromine-containing solid is selected from the group consisting of the alkali metal salt of anhydrous N-bromosulfamate and the earth alkali metal salt of anhydrous N-bromosulfamate.

75. A method of preparing a bromine-containing solid in equilibrium with its saturated solution, comprising:

- a. combining the source of bromide ions, a complexing agent, a first alkaline source, and a solid, organic halogenating agent;
- b. conducting a solid-liquid separation;
- c. adding a second alkaline source;
- d. promoting crystallization of a bromine-containing solid; and
- e. recovering a slurry of said bromine-containing solid in equilibrium with its saturated solution.

76. The method of Claim 75, wherein said bromine-containing solid is selected from the group consisting of the alkali metal salt of hydrated N-bromosulfamate and the earth alkali metal salt of hydrated N-bromosulfamate.
77. A bromine-containing liquid, having an active ingredient content of at least about 18% as Br₂ (8% as Cl₂).
78. The bromine-containing liquid of Claim 77, prepared to contain between zero and about one mole of dissolved halide ion salts per mole of active halogen.
79. The bromine-containing liquid of Claim 78, developing no solid precipitates for up to 44 days when stored at about 120° F to about 130° F.
80. The bromine-containing liquid of Claim 78, having an active ingredient half-life of at least about 58 days at about 125° F.
81. A bromine-containing liquid, prepared to contain between zero and about one mole of dissolved halide ion salts per mole of active halogen.
82. The bromine-containing liquid of Claim 81, prepared to contain less than one mole of dissolved halide ion salts per mole of active halogen.
83. A bromine- and chlorine-containing liquid, having a total halogen content of at least about 2.25% when expressed as Br₂ (1% when expressed as Cl₂).
84. The liquid of Claim 83, having less than one mole of dissolved halide ion salts per mole of active halogen.
85. The liquid of Claim 83, developing no solid precipitates for up to 34 days when stored at about 120° F.
86. A solid composition of matter, comprising the salt of N-bromosulfamate.

87. The solid composition of Claim 86, wherein said N-bromosulfamate is hydrated N-bromosulfamate.
88. The solid composition of Claim 87, wherein said salt is selected from the group consisting of alkali metal and earth alkali metal.
- 5 89. The solid composition of Claim 88, wherein said solid is in equilibrium with its saturated solution as a slurry.
90. The solid composition of Claim 86, wherein said N-bromosulfamate is anhydrous N-bromosulfamate.
91. The solid composition of Claim 90, wherein said salt is selected from the group
10 consisting of alkali metal and earth alkali metal.
92. The solid composition of Claim 91, wherein said solid is extremely stable if kept dry.
93. A method for microbiological control in industrial water systems, comprising the addition to the water of the bromine-containing liquid composition of
15 Claim 77.
94. A method for microbiological control in industrial water systems, comprising the addition to the water of the mixed halogen liquid composition of Claim 83.
95. A method for microbiological control in industrial water systems, comprising the
20 addition to the water of the solid bromine-containing composition of Claim 86.

96. A method for microbiological control in residential water systems, comprising the addition to the water of the bromine-containing liquid composition of Claim 77.
97. A method for microbiological control in residential water systems, comprising the addition to the water of the mixed halogen liquid composition of Claim 83.
98. A method for microbiological control in residential water systems, comprising the addition to the water of the solid bromine-containing composition of Claim 86.
99. A bromine-containing liquid composition for microbiological control in industrial water systems, comprising the composition of Claim 77.
100. A bromine- and chlorine-containing liquid for microbiological control in industrial water systems, comprising the composition of Claim 83.
101. A solid composition for microbiological control in industrial water systems, comprising the composition of Claim 86.
102. A bromine-containing liquid composition for microbiological control in residential water systems, comprising the composition of Claim 77.
103. A bromine- and chlorine-containing liquid for microbiological control in residential water systems, comprising the composition of Claim 83.
104. A solid composition for microbiological control in residential water systems, comprising the composition of Claim 86.